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(54) FLAME-RESISTANT RESIN COMPOSITION

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(72) Inventor: Teruo Ozaki  
6-5, Sawazenshin-cho, Uogyo-shi

(71) Applicant: Nihon Carbide Co., Ltd.  
3-1, Marunouchi 3 chome, Chiyoda-ku, Tokyo-to

(74) Agent: Akira Aoki, Patent Attorney and 3 others

## Specification

### 1. Title of the Invention

#### FLAME-RESISTANT RESIN COMPOSITION

### 2. Scope of Patent Claim

1. A flame-resistant resin composition comprising (i) at least one resin component selected from the group consisting of phenol, amino, epoxy, and unsaturated polyester thermosetting resin as well as polyolefin, styrene vinyl acetate, acrylic, polyether, polycarbonate, polyester and urethane thermoplastic resin and (ii) melamine cyanurate.

2. The composition in claim 1, where the above-mentioned resin component is at least one resin selected from the group of amino thermosetting resin, acrylic thermoplastic resin, and polyether thermoplastic resin.

3. The composition in claim 1, where the above-mentioned resin component is polyether thermoplastic resin.

4. The composition in claim 1, 2, or 3, wherein the amount of above-mentioned melamine cyanurate is 3 to 50 parts by weight per 100 parts by weight of the above-mentioned resin component.

### 3. Detailed Description of the Invention

The present invention pertains to a flame-resistant resin composition.

It is a known fact that various types of plastics are used in many fields because of their excellent mechanical properties, electrical properties, chemical resistance, thermal properties, physical properties or workability, etc. For instance, they are used as important industrial materials in the fields of electrical components, various interior materials, fibers, paints, adhesives, miscellaneous products, etc. However, many plastics have a disadvantage in that they readily catch fire. Moreover, there is a rising demand for flame-resistant plastics as a result of their increased use and the strengthening of various legal restrictions.

Various flame retarders have been developed to meet this demand. Of these, antimony compounds and halogen compounds or mixtures of the same are generally used because there are relatively few restrictions in terms of the plastics they can be used with and their flame-retarding activity is high. Nevertheless, the flame retarders themselves are toxic or they give off toxic gases when they decompose, or they are corrosive to metals, and therefore, resin compositions containing large amounts of these flame retarders pose problems in terms of safety and environmental pollution.

In contrast to this, nitrogen compounds such as melamine and cyanuric acid (or isocyanuric acid), etc., have flame-retarding activity and pose little risk in terms of the above-mentioned environmental pollution, but melamine has disadvantages in that it plates out on to the mold surface when the resin composition is molded, it is difficult to uniformly disperse large amounts of melamine in the polymer, and further, melamine often induces a decrease in plastic properties. On the other hand, although the flame-retarding activity of

cyanuric acid is relatively strong, it has a disadvantage in that it moves to the surface with long-term storage and so-called blooming occurs. This prior art is disclosed in, for instance, Japanese Kokai Patent No. Sho 50(1975)-105,744, Japanese Kokai Patent No. Sho 51(1976)-54,655, etc.

The inventors performed intense studies to solve the above-mentioned problems with conventional flame retarders for plastics and as a result, they successfully completed the present invention upon discovering that excellent flame-retarding activity is realized and there are no problems in terms of safety, environmental pollution or workability when melamine cyanurate, which is the reaction product of melamine and cyanuric acid, is mixed with a specific resin as the flame retarder.

The flame-resistant resin composition of the present invention comprises (i) at least one resin component selected from the group consisting of phenol, amino, epoxy, and unsaturated polyester thermosetting resin as well as polyolefin, styrene, vinyl acetate, acrylic, polyether, polycarbonate, polyester and urethane thermoplastic resin and (ii) melamine cyanurate.

One or more phenol thermosetting resins, such as phenol, cresol, thermosetting resin obtained by addition condensation of phenol such as xylenol, etc., and formaldehyde, phenol-furfural resin, etc., amino thermosetting resin, such as melamine resin, melamine phenol resin, aniline resin, sulfonamide resin, benzoguanamine resin, acetoguanamine resin, etc.; epoxy thermosetting resin; polyolefin thermoplastic resin, such as polyethylene, polypropylene, polybutylene, etc.; styrene thermoplastic resin, such as polystyrene, ABS resin,

high-impact polystyrene, AS resin, etc.; vinyl acetate thermoplastic resin; acrylic thermoplastic resin, such as polyacrylate, polymethacrylate, polyacrylonitrile, etc.; polyether thermoplastic resin, such as polyformaldehyde, polyacetaldehyde, polyethylene oxide, polypropylene oxide, polycyclooxabutane, polytetrahydrofuran, polycarbonate thermoplastic resin, polyester thermoplastic resin; or urethane resin is used as the resin component mixed with the flame-resistant resin composition of the present invention. The preferred resins are amino thermosetting resin, acrylic thermoplastic resin and polyether thermoplastic resin, and polyether thermoplastic resin is particularly preferred.

The flame retarder mixed with the flame-resistant resin composition of the present invention, melamine cyanurate, is a salt that is produced by the reaction between melamine and cyanuric acid and is produced, for instance, by adding an aqueous cyanuric acid suspension to an aqueous melamine suspension that has been heated and heating to a temperature of 90 - 100°C. The following structure, for instance, appears to be the structure of the salt that is produced:

The composition ratio of the flame-resistant resin composition of the present invention is preferably 3 to 50 parts by weight, particularly 5 to 30 parts by weight of melamine cyanurate, per 100 parts by weight of the above-

mentioned resin component. If there is less than 3 parts by weight of melamine cyanurate, flame-retarding activity will be insufficient, while if it exceeds 50 parts by weight, a plating out and poor dispersion will be the undesirable results. The flame-resistant resin composition of the present invention may contain additives generally used with resins, including curing agents and curing promoters in the case of thermosetting resins, fillers, pigments, dyes, ultraviolet radiation absorbing agents, stabilizers, lubricants, plasticizers, foaming agents, antioxidants, charge inhibitors, antifogging agents, etc., in addition to the above-mentioned resin component and melamine cyanurate. These can be mixed using a general resin mixing means, such as a Henschel mixer, ribbon blender, extrusion kneader, Banbury mixer, ball mill, co-kneader, mixing roll, etc.

When melamine cyanurate is mixed with the above-mentioned resin in accordance with the present invention, results are produced that are much better than when melamine or cyanuric acid (or isocyanuric acid) or their mixture is used. The main results when melamine cyanurate is used are the very strong flame-retarding activity and the fact that plating out on the mold will not occur, there is uniform dispersion in the plastic, there is no so-called blooming with long-term storage, etc. That is, when melamine is used, there are problems in that sublimation of the melamine occurs during molding and so-called plating out where the melamine deposits on the mold or rolls occurs leading to contamination of the molded article or poor mold release. Moreover, when cyanuric acid is used, there is a disadvantage in that there is a reduction in the properties of the molded product and blooming is marked. These problems are

not solved when the two are used concomitantly. However, when melamine cyanurate, which is a salt of melamine and cyanuric acid, is mixed with the above-mentioned resin, the above-mentioned problems are solved, there is almost no plating out on the mold or roll surface, and blooming is improved

Examples of the present invention will now be described:

#### Example 1

1, 3, 10, 30, 50 and 60 parts by weight of melamine cyanurate were added to 100 parts by weight of Duracon M-25-01 (polyacetal resin, Polyplastic Co., Ltd.) and kneaded for 10 minutes at 150°C using rolls with a diameter of 6 inches. This bank fluid was pressed at 200°C to obtain pressed sheets with a thickness of 0.5 mm and the roll surface, the surface of the pressing sheet and the appearance of the pressed sheets that were obtained were checked to evaluate dispersion of the melamine cyanurate, plating out and blooming after 1 month. The results are shown in Table 1.

Moreover, a 10 mm x 100 mm test piece was cut from the pressed sheet that was made and the burned state and state after removing the gas burner were macroscopically checked when the test pieces was brought into contact with a gas burner flame for 10 seconds. The results in Table 1 were obtained.

By way of comparison, the same tests were performed with Duracon M-25-01 only and when 30 parts by weight of melamine, 30 parts by weight of cyanuric acid, and 30 parts by weight of a mixture of equal amounts of melamine and cyanuric acid were added in place of melamine cyanurate. The results are shown in Table 1.

Table 1.

RUN No.	Flame retarder		Flammability			Plating out onto roll or pressing-plate surface	Blooming of sheets
	Type	Amount (PHR)	Flame resistance	Molten drops	Smoking		
1*	Melamine cyanurate	1	Δ	x	O	no	no
2	"	3	O	Δ	O	"	"
3	"	10	(O)	O	O	"	"
4	"	30	(O)	O	O	"	"
5	"	50	(O)	O	O	"	"
6	"	60	(O)	O	O	somewhat	yes
7*	None	--	x	x	O	no	no
8*	Melamine	30	Δ	Δ	O	yes	yes
9*	Cyanuric acid	30	O	Δ	O	yes	yes
10*	Melamine + cyanuric acid	30	O	Δ	O	yes	yes

## Footnotes:

\*Comparative Example

\*\*Macroscopic evaluation

(O)	Excellent
O	Good
Δ	Somewhat good
X	Poor

## Examples 2 to 12

The same tests as in Example 1 were performed when 30 PHR melamine cyanurate was added to the various resins in Table 2 instead of the polyacetal resin in Example 1 and when it was not. The results are shown in Table 2.

Furthermore, the test piece was made from a sheet that was produced by pressing roll-kneaded sheets or bank fluid from roll kneading when roll sheets were not obtained because [the product became] sticky when heated. Moreover, pressed sheets with a thickness of 0.5 mm were obtained by kneading and dispersing melamine cyanurate in thermosetting plastics at a roll temperature of



50°C or lower and then adding curing agent, promoters, etc., immediately before pressing for 10 minutes at 170 to 200°C. Moreover, the surface of the pressed sheet at this time was checked.

Table 2.

Example No.	Resin	Roll kneading		Amount of melamine cyanurate added (PHR)	Flammability *1 *2				Plating out onto roll or pressing-plate surface	Sheet blooming
		Temperature (°C)	Time (minutes)		Flame resistance	Molten drops	Smoking			
2	Polyethylene (Mitsubishi Kasei, Nobatek J 5050)	140	10	30/--	Δ/x	Δ/x		o/o		o/o
3	ABS resin (Mitsubishi Rayon Daiyapet HF-5)	140	10	30/--	Δ/x		o/o	Δ/x		o/o
4	Ethylene-vinyl acetate copolymer (Mitsui Sekiyu Kagaku Evaprex 250)	50	10	30/--	Δ/x	Δ/x		o/Δ		o/o
5	Polymethacrylate (Mitsubishi Rayon Acripet MD)	150	10	30/--	o/x		o/x		o/o	o/o
6	Polycarbonate (Mitsubishi Gas Chemical Yupiron MS-3000)	170	10	30/--	o/Δ	Δ/x		o/Δ		o/o
7	Polyurethane (Japan Polyurethane Paraprene 25S-M)	160	10	30/--	o/Δ		o/x		o/o	o/o
8	Polyester (Toyobo Perprene P40H)	120	10	30/--	o/x	Δ/x		o/Δ		o/o
9	Phenol resin (Mitsui Toa Novolak 1000)	40	10	30/--	(o)/Δ	(o)/(o)		o/Δ		o/o
10	Melamine resin (Japan Carbide Nicalazon S-176)	50	10	30/--	(o)/o	(o)/(o)		o/o		o/o

11	Epoxy resin (Shell Epicoat 828)	40	10	30/--	o/Δ	(o)/(o)	o/Δ	o/o	o/o
12	Unsaturated polyester resin (Hitachi Kasei Polyset 650ML)	40	10	30/--	(o)/Δ	(o)/(o)	o/o	o/o	o/o

\*1O represents the case where 30 parts by weight of melamine cyanurate were added and X represents the case where none was added.

<sup>2</sup>Refer to the footnote of Table 1 for interpretation of symbols.